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(19)

(72) Inventors HANNES FISCHER and HANS-DIETER MUMMENTHEY

(54) POWDER COATING COMPOSITION

(71) We, HOECHST AKTIENGESELLSCHAFT, a Body Corporate organised under the laws of the Federal Republic of Germany, of 6230 Frankfurt/Main 80 Germany, do hereby declare the invention, for which we pray that a patent may be granted to us, and the method by which it is to be performed, to be particularly described in and by the following statement:—

This invention relates to powder coating compositions and to a process for the preparation thereof.

Heat-setting synthetic resins and synthetic resin mixtures are particularly suitable as binding agents for the preparation of powder coating compositions, for application for example by the whirl sinter process or the electrostatic powder coating process, since they yield coatings which are generally resistant to chemicals and to mechanical stresses. In those cases where the UV-resistance of the coating is of secondary importance, epoxy resin powders are preferably used, whereas to prepare weather-resistant coatings, polyester resins are chiefly used. However, the high hardening temperatures required by the latter, generally above 200°C, are a disadvantage.

It has been proposed to use mixtures of polyesters and epoxy group containing compounds in an attempt to combine the properties of these two classes of compounds in a single powder coating product. Thus, the mixing of certain polyesters containing free carboxyl groups with triglycidyl isocyanurate in the presence of organic acid anhydrides and the use of the products thus obtained as binding agents for powder resins have already been described. It has also been proposed to use mixtures of polycarboxylic acid anhydrides and tertiary amines or quaternary ammonium compounds as hardening accelerators for mixtures of polyesters containing carboxyl groups and epoxy resins, and to use the products as synthetic resins for preparing hardenable moulding materials. It has further been proposed to prepare crystallisable products suitable as binding agents for powder coating compositions by mixing certain acid group containing polyesters with epoxy resins. Finally powder containing compositions with allegedly good properties have been described containing mixtures of polyesters containing free carboxyl groups and glycidyl esters of polycarboxylic acids.

A disadvantage of the previously described resin combinations is that the individual components tend to react with one another during the preparation of the powder coating composition, which is usually performed according to a technically conventional process by mixing the components of the composition in a melt at temperatures of from 80 to 140°C, preferably from 100 to 120°C. Thus a partial reaction may take place between the acidic polyester resins and the compounds containing the epoxy groups of the afore-described powder lacquer binding agents leading to premature partial cross-linking and therefore the coatings prepared from such powder lacquer binding agents may be of poor quality.

According to the present invention we now provide a powder coating composition comprising a) an oligomeric ester or a polyester containing free hydroxyl groups but substantially no free carboxyl groups, b) a compound containing at least 2 epoxy groups and c) a nitrogenous base or thermally decomposable salt thereof.

The nitrogenous base or thermally decomposable salt thereof acts as a hardening catalyst and ensures that substantially no reaction takes place under the mixing conditions between the hydroxyl groups of the ester or polyester and the epoxy groups. Thus undesirable cross-linking during the preparation of the coating composition is substantially reduced or eliminated, and the subsequent processability of the coating composition is ensured.

The ester or polyester component a) is preferably derived from at least one substituted or unsubstituted aromatic di- to tetracarboxylic

acid or saturated or olefinically unsaturated aliphatic di- to tetracarboxylic acid (e.g. a hydrogenation product of one of the aforementioned aromatic acids) and at least one saturated or olefinically unsaturated aliphatic (including cycloaliphatic) di- to tetrahydric alcohol containing from 2 to 15 carbon atoms. Particularly preferred are esters and polyesters from carboxylic acids containing from 4 to 10 carbon atoms, for example ortho-, iso- and terephthalic acids and their hydrogenation and/or substitution products, trimellitic acid, pyromellitic acid, maleic, fumaric, itaconic, adipic, glutaric and succinic acid, and from polyhydric alcohols with 2 to 15 carbon atoms, such as ethanediol, the propane-, butane-, pentane- and hexanediols, dimethylolcyclohexane, bis (4-hydroxy-cyclohexyl) methane and propane, Δ-2,3-butene-1,4-diol, glycerol, trimethylolethane and trimethylolpropane and pentaerythritol.

The content of dicarboxylic acid in the ester or polyester a) is preferably at least 70, more preferably at least 85 and particularly at least 90 equivalent % (based on the total carboxylic acid content) and the content of dihydric alcohols is preferably at least 40, more preferably at least 50 equivalent % (based on the total alcohol content).

Polyesters containing at least one trifunctional acid or alcohol component are preferred. Higher functional acids or alcohols may also be incorporated but it is preferred that the proportion of carboxylic acids containing 3 or more carboxyl groups and of tri- or higher functional alcohols should not be more than 10 equivalent % (based respectively on the total acid or alcohol content). The total proportion of bifunctional components, i.e. dicarboxylic acids and dihydric alcohols, is desirably at least 50, preferably at least 65 equivalent % based on the total alcohol and acid content in the ester. The ratio of OH groups to COOH groups in the starting mixture used to prepare the ester or polyester is appropriately

(50 to 80):(50 to 20),

to obtain the desired hydroxyl number in the ester component.

The ester used for preparing the powder coating compositions according to the invention may if desired contain at least one monocarboxylic acid component, preferably a monocarboxylic acid containing at least 7 carbon atoms, for example, benzoic acid, *p*-tert-butylbenzoic acid, an aliphatic (including cycloaliphatic) saturated or olefinically unsaturated monocarboxylic acid such as a fatty acid of a naturally occurring oil e.g. linseed oil, soya bean oil, castor oil or coconut oil, or dehydrated castor oil fatty acid, stearic acid or oleic acid. The ester may if desired also contain at least one monohydric alcohol component, preferably an aliphatic or aromatic alcohol containing from 2 to 18 carbon atoms, for example ethanol, propanol, butanol, pentanol, hexanol, cyclohexanol, or a monohydric alcohol with a longer carbon chain, such as lauryl alcohol or oleyl alcohol.

Terephthalic acid polyesters are preferred.

The ester component a) must be substantially free of carboxyl groups: its acid number, measured according to DIN standard 53402, is desirably 5 or less, preferably 3 or less. The hydroxyl number of the ester components, determined according to DIN standard 53240, is advantageously from 50 to 250, preferably from 100 to 200, and particularly from 130 to 190. The melting points of the esters, measured according to the capillary method, are generally from 60 to 90, preferably from 65 to 80°C and they have a melting viscosity which is generally from 100 to 1400 poise at 130°C. However, esters with higher melting viscosities, for example 1400 to 3000 poise at 130°C, may also be used. A low melting viscosity is particularly advantageous since then processing of the ester component into the powder coating composition is easier.

The oligomeric ester or polyester a) may be prepared from its acid and alcohol components by methods known *per se* for polycondensation resins, e.g. by condensation in a melt, by esterification in an azeotropic solvent mixture with recycling of the solvent mixture, or by ester interchange with high molecular weight polyesters, advantageously under a protective gas atmosphere, e.g. under nitrogen or carbon dioxide.

The esterification is generally performed at temperatures from 200 to 250°C, the final stages of the reaction being carried out under a reduced pressure to raise the melting point of the ester product to the desired level. However, working under reduced pressure is preferably kept to a minimum.

The epoxy group containing compound b) has on average at least two epoxy groups per molecule and produces in combination with the ester component a) and base c) a pulverisable, non-tacking binder for the powder coating composition of the invention. The epoxy compound is preferably triglycidyl isocyanurate but others which we have used include those derived from novolaks, (e.g. diphenylol propane and di- and triphenylolmethane), and epichlorohydrin, and condensation products prepared from aryloxychlorohydrins and formaldehyde, and optionally chlorine-free hydroxyl group containing phenol ethers, with subsequent elimination of hydrogen chloride. Bifunctional epoxy resins, for example the bisglycidyl ethers of diphenylolpropane and diphenylolmethane, may advantageously also be used as component b) of the powder composition according to the invention and also 3,4-epoxy-6-cyclohexylmethyl 3′,4′-epoxy-6′-methyl-cyclohexanecarboxylate, N,N-diglycidyl-dimethyl-hydantoin, 2,2-bis (epoxycyclohexane) propane and other cycloaliphatic epoxy compounds. Other bi- and

higher functional epoxy resins may also be used including epoxy resins of higher molecucular weight than those above specified. Moreover, halogen-substituted, e.g. chlorine-containing epoxy compounds, particularly epoxy resins, and internal esters of the abovementioned epoxy resins, may be used. It is also possible to use mixtures of epoxy compounds as component b). The average number of epoxy groups per molecule of polyepoxy compound is preferably at least 3.

By adding the nitrogenous base c) or a thermally decomposable salt thereof, the temperature required for substantially complete cleavage of the epoxy groups of component b) may in general be reduced to temperatures of from 150 to 200°C, preferably from 160 to 200°C and by appropriate choice of catalyst c) the above temperature may be adjusted to the desired figure. On the other hand, the basic catalyst c) used according to the invention only accelerates the reaction between epoxy and hydroxyl groups to a small extent at temperatures below 140°C. Thus the preparation and manufacture of the powder coating compositions according to the invention may be carried out without any substantial cross-linking of the hydroxyl groups of the ester a) and epoxy groups of component b). The catalyst c) may if desired be a salt of a nitrogenous base which is decomposable at elevated temperature, generally at a temperature of 150°C or higher, to yield the free base and a free acid. Thus the effectiveness of the accelerator component c) may be restricted to a particular temperature range, for example temperatures above 150°C, preferably from 160 to 200°C.

Preferred nitrogenous bases and salts thereof which may be used as component c) are in particular amines including aliphatic (including cycloaliphatic and heterocyclic) and aromatic (including aromatic heterocyclic) amines, particularly amines containing both aliphatic and aromatic groups, the said amines being optionally substituted by hydroxyl or alkyl groups or by halogen atoms and their salts with a mineral acid, preferably their hydrochloric acid salts and salts with short-chained aliphatic monocarboxylic acids having up to 6 carbon atoms. Secondary and particularly tertiary amines are preferably used and also quaternary ammonium salts, e.g. tetramethyl, tetraethyl and tetrabutyl ammonium chloride, and quaternary ammonium hydroxides. Thus for example N - methylbenzylamine, N - methylaniline, dimethylamine, dibutylamine, diethanolamine, 4,4'-diaminodiphenylmethane, diazabicyclooctane, trimethylamine, triethylamine, tributylamine, tribenzylamnie, triethanolamine, dimethylaniline, dimethylbenzylamine, tris (dimethylaminomethyl) phenol, and various pyridines and morpholines may be used. Suitably component c) may be a tertiary aralkylamine or a thermally decomposable salt thereof.

The power coating compositions according to the invention generally contain from 5 to 30%, preferably from 7 to 25% and in particular from 10 to 20% by weight [referred to the weight of ester component a)] of the epoxy component b) and from 0.2 to 4, preferably from 0.5 to 2% by weight [again referred to the weight of ester component a)] of the nitrogenous base or thermally decomposable salt thereof.

According to a further feature of the invention there is provided a process for the preparation of a powder coating composition which comprises mixing in a melt a) an oligomeric ester or a polyester containing free hydroxyl groups but substantially no carboxyl groups, b) a compound containing at least 2 epoxy groups and c) a nitrogenous base or thermally decomposable salt thereof, solidifying the molten mixture and comminuting the solid mass.

The mixing process is generally carried out at a temperature of 80 to 130°C, preferably from 80 to 110°C and particularly from 80 to 95°C, until a homogeneous solution or dispersion is produced. The mixing is conveniently performed in a kneader, mixer or extruder.

If desired one or more further components selected from pigments, fillers and other additives, e.g. levelling agents, may be incorporated into the melt and evenly distributed therein prior to, during or after mixing. The proportion of pigments is preferably in the range 30 to 50% by weight, fillers 20 to 40, more preferably 25 to 35% by weight, and levelling agents 0.2 to 5% by weight, in each case referred to the total weight of components a), b) and c). In this way, powder coating compositions which are ready for use may be prepared in one operation.

On cooling the mixture solidifies to a solid mass either as a single solid body or as granules. This mass may then be comminuted e.g. by grinding and sieving, into a freely flowing powder which does not agglomerate even on storage for a fairly long time.

The powder coating compositions may be applied to suitable manufactured articles, e.g. by whirl sintering or by the electrostatic powder spray process, and then hardened, by heating generally at 160 to 200°C, preferably 170 to 190°C, for example for 30 minutes. In general even, glossy coatings with good mechanical and chemical resistance and with good weather resistance are produced.

The following Examples serve to illustrate the preparation of the powder coating compositions according to the invention and of starting materials therefor. In the Examples, T means parts by weight and all percentages are by weight. The melting viscosities were determined at 130°C unless otherwise stated.

The following Examples A to C illustrate

the preparation of polyesters used as ingredients in the powder compositions of Examples 1 to 4 and the Comparison Example which follow.

Example A.

Polyester A

1460 g of trimethylolpropane (10.9 mol) and 3770 g of polyethylene glycol terephthalate (19.6 mol) were heated at 260°C in a CO_2 atmosphere until a clear solution was formed. The mixture was refluxed for 3 hours at 240°C, and then ethylene glycol was distilled off, under a vacuum of 16 mm Hg as the temperature fell, until a reaction product having a softening point of 70 to 75°C was obtained. The polyester obtained had a hydroxy number of 175±5 (according to DIN standard 53 240), an acid number of less than 5 and a melting viscosity of 2400 poise.

Example B.

Polyester B

2350 of isophthalic acid (19.6 mol), 1460 g of trimethylolpropane (10.9 mol) and 675 g of ethylene glycol (10.9 mol) were heated at 220°C, in a CO_2 atmosphere with recycling distillation until a clear solution formed. Subsequently ethylene glycol was distilled off at 200 to 220°C, finally reducing the pressure to 16 mm Hg. A polyester with a softening point of 71 to 72°C, a hydroxy number of 180+5 (according to DIN standard 53240), an acid number of less than 5 and a melting viscosity of 2200 P was obtained.

Example C.

Polyester C (comparison)

A known polyester resin was prepared from a diether of ethylene glycol and diphenylolpropane, the glycidyl esters of a mixture of branched fatty acids containing 9 to 11 C-atoms, terephthalic acid and trimellitic acid according to Example 1 of German offenlegungsschrift 2 163 962. This polyester had the following characteristics: softening point: 93°C; acid number (according to DIN standard 53402): 57; melting viscosity: 2250 P.

Example 1.

Preparation of powder coating composition containing Polyester A

A homogeneous, highly-viscous melt was prepared from polyester A, triglycidyl isocyanurate, N,N - dimethylbenzylamine hydrochloride, poly-(ethylhexyl methacrylate) and titanium dioxide by mixing the various components in the proportions indicated in Table I which follows at 90 to 100°C in a heated mixer provided with Z-shaped kneading elements. The melt is cooled to room temperature and is then broken into pieces which are ground and then sieved to a particle size of less than 100 μm.

Example 2.

Preparation of powder coating composition containing Polyester B

A powder composition of maximum particle size 100 μm is prepared from polyester B analogously to Example 1.

Example 3.

Preparation of powder coating composition containing Polyester C

A powder composition of maximum particle size 100 μm is prepared from polyester A analogously to Example 1 but containing the free base N,N - dimethylbenzylamine as catalyst.

Example 4.

Preparation of powder coating composition containing polyester A

A powder coating composition was prepared from 300 g polyester A, 80 g of an epoxy resin prepared from diphenylolpropane and epichlorohydrin (epoxy equivalent weight: 450 to 525; melting point: 60 to 70°C), 3g of N,N - dimethylbenzylamine hydrochloride, 33% (based on the resin/catalyst mixture) titanium dioxide and 3 g of flow agent by the method described in Example 1.

Comparison Example.

A powder composition is prepared from polyester C, triglycidyl isocyanurate; titanium dioxide and tris(dimethylaminomethyl) phenol in the proportions indicated in Table 1 analogously to Example 1.

TABLE 1

Example	Polyester	% Epoxy Compound (1)	%TiO$_2$ (2)	% Catalyst (1)	Levelling % agent (1) (3)
1	A	14 (4)	33	1.5 I	1
2	B	14 (4)	33	1.5 I	1
3	A	14 (4)	33	0.5 III	1
4	A	26.7(5)	33	1 I	1
Comparison	C	13.7(4)	33	0.3 II	1

Explanations:
(1) based on polyester
(2) based on total binding agent
(3) poly-(ethylhexyl methacrylate)
(4) triglycidyl isocyanurate
(5) epoxy resin
I dimethylbenzylamine HCl
II tris (dimethylaminomethyl) phenol
III dimethylbenzylamine

Technical application test.

The powder compositions prepared in Examples 1 to 4 and the comparison Example were electrostatically sprayed on to steel sheets (Bonder 130) in a film thickness of 60 to 70 μm, and then hardened for 30 minutes at 190°C. The properties of the coatings prepared therefrom are assembled in the following Table 2. In the chemical resistance and film flow tests evaluation is according to DIN standard 53230 (0=best value, 5= worst value):

TABLE 2

Example	Resistance to acetone	Resistance to xylene	Erichsen cupping (mm)	Film flow
1	0–1	0	9.3	3
2	0	0	7.8	3
3	2	0	10.3	2
4	3	1	6.0	2.5
Comparison	5	2	0.2	5

Discussion of results.

As indicated in Table 2 the coatings prepared from the coating compositions according to the invention prepared in Examples 1 to 4 are superior to the coating prepared from the known composition of the Comparison Example with respect to chemical resistance and film flow.

WHAT WE CLAIM IS:—

1. A powder coating composition comprising a) an oligomeric ester or a polyester containing free hydroxyl groups but substantially no free carboxyl groups, b) a compound containing at least 2 epoxy groups and c) a nitrogenous base or thermally decomposable salt thereof.

2. A composition as claimed in claim 1 wherein the ester component a) has a hydroxyl number of from 50 to 250.

3. A composition as claimed in claim 1 wherein the ester component a) has a hydroxyl number of from 100 to 200.

4. A composition as claimed in claim 1 wherein the ester component a) has a hydroxyl number of from 130 to 190.

5. A composition as claimed in any of the

preceding claims wherein the ester component a) has an acid number of 5 or less.

6. A composition as claimed in any of the preceding claims wherein the ester component a) has an acid number of 3 or less.

7. A composition as claimed in any of the preceding claims wherein the ester component a) is derived from at least one substituted or unsubstituted aromatic di- to tetracarboxylic acid or saturated or olefinically unsaturated aliphatic di- to tetracarboxylic acid and at least one saturated or olefinically unsaturated aliphatic (including cycloaliphatic) dihydric to tetrahydric alcohol containing from 2 to 15 carbon atoms.

8. A composition as claimed in claim 7 wherein in the ester component a) the content of dicarboxylic acids is at least 70 equivalent % (referred to the total carboxylic acid content) and the content of dihydric alcohol is at least 40 equivalent % (referred to the total alcohol content).

9. A composition as claimed in claim 8 wherein in the ester component a) the content of dicarboxylic acids is at least 85 equivalent % (referred to the total carboxylic acid content) and the content of dihydric alcohols is at least 50 equivalent % (referred to the total alcohol content).

10. A composition as claimed in any of the preceding claims wherein the ester component a) contains at least one tricarboxylic acid or trihydric alcohol component.

11. A composition as claimed in any of the preceding claims wherein the ester component a) contains at least one mono-carboxylic acid or monohydric alcohol component.

12. A composition as claimed in claim 11 wherein the ester component a) contains at least one monocarboxylic acid selected from benzoic acid, *p*-tert.-butylbenzoic acid, saturated and olefinically unsaturated fatty acids from naturally occurring oils and stearic acid.

13. A composition as claimed in claim 11 wherein the ester component a) contains at least one saturated or olefinically unsaturated aliphatic (including cycloaliphatic) monohydric alcohol containing from 2 to 18 carbon atoms.

14. A composition as claimed in any of the preceding claims wherein the ester component a) is a terephthalic acid polyester.

15. A composition as claimed in any of the preceding claims wherein the ester component a) has a melting point of from 60 to 90°C and a melting viscosity of from 100 to 3000 P/130°C.

16. A composition as claimed in claim 15 wherein the ester component a) has a melting point of from 65 to 80°C and a melting viscosity of from 100 to 1400 P/130°C.

17. A composition as claimed in any of the preceding claims wherein the ester component a) is prepared by condensation at 200 to 250°C and towards the end of the condensation at a pressure below atmospheric presure.

18. A composition as claimed in any of the preceding claims wherein the epoxy compound b) is an addition product of epichlorohydrin to a di- or triphenylolalkane, a condensation product of an aryloxychlorohydrin and formaldehyde by elimination of hydrogen chloride, triglycidyl isocyanurate or a cycloaliphatic polyepoxy compound, or a halogenation or esterification product thereof.

19. A composition as claimed in claim 18 wherein the epoxy compound b) is a condensation product of an aryloxychlorohydrin, formaldehyde and a chlorine-free phenol ether.

20. A composition as claimed in any of the preceding claims wherein the epoxy compound b) contains at least 3 epoxy groups per molecule.

21. A composition as claimed in any of the preceding claims wherein component c) is a secondary or tertiary amine or a quaternary ammonium salt or a quaternary ammonium hydroxide.

22. A composition as claimed in claim 21 wherein component c) is an aliphatic (including cycloaliphatic), aromatic or heterocyclic amine being unsubstituted or substituted by at least one hydroxyl, alkyl or halogen substituent or a salt thereof with a mineral acid or a short-chained mono-carboxylic acid having up to 6 carbon atoms.

23. A composition as claimed in claim 22 wherein component c) is a tertiary aralkylamine or a thermally decomposable salt thereof.

24. A composition as claimed in claim 23 wherein component c) is dimethylbenzylamine or tris(dimethylaminomethyl) phenol or a thermally decomposable salt thereof.

25. A composition as claimed in any of the preceding claims wherein component c) is a salt of a nitrogenous base which is decomposable at a temperature of 150°C or higher.

26. A composition as claimed in any of the preceding claims containing from 5 to 30% by weight [referred to the weight of ester a)] of the epoxy compound b).

27. A composition as claimed in claim 26 containing from 7 to 25% by weight [referred to the weight of ester a)] of the epoxy compound b).

28. A composition as claimed in claim 26 containing from 10 to 20% by weight [referred to the weight of ester a)] of the epoxy compound b).

29. A composition as claimed in any of the preceding claims containing from 0.2 to 4% by weight [referred to the weight of ester a)] of component c).

30. A composition as claimed in claim 29 containing from 0.5 to 2% by weight [referred to the weight of ester a)] of component c).

31. A composition as claimed in any of the preceding claims which additionally contains

a pigment, filler or levelling agent.

32. A composition according to claim 1 substantially as herein described.

33. A composition according to claim 1 substantially as herein described with reference to Examples 1 to 4.

34. A process for the preparation of a powder coating composition which comprises mixing in a melt a) an oligomeric ester or a polyester containing free hydroxyl groups but substantially no carboxyl groups, b) a compound containing at least 2 epoxy groups and c) a nitrogenous base or thermally decomposable salt thereof, solidifying the molten mixture and comminuting the solid mass.

35. A process as claimed in claim 34 wherein the mixing is performed at temperatures of from 80 to 130°C.

36. A process as claimed in claim 34 wherein the mixing is performed at temperatures of from 80 to 110°C.

37. A process as claimed in claim 36 wherein the mixing is performed at a temperature of from 80 to 95°C.

38. A process as claimed in any of claims 34 to 37 wherein the mixing is performed in a kneader, mixer or extruder.

39. A process as claimed in any of claims 34 to 38 wherein the solid mass is comminuted by breaking it up into lumps and then grinding and finally sieving.

40. A process as claimed in any of claims 34 to 38 wherein at least one further ingredient selected from pigments, fillers and levelling agents is incorporated into the melt.

41. A process as claimed in any of claims 34 to 40 substantially as herein described.

42. A process as claimed in any of claims 34 to 40 substantially as herein described in Examples 1 to 4.

43. A powder coating composition whenever prepared by a process as claimed in any of claims 34 to 42.

For the Applicants,
FRANK B. DEHN & CO.,
Imperial House,
15—19 Kingsway,
London, WC2B 6UZ.

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